

#2
Nickie & Susan
5-10-96

EXAMINER :

ART UNIT :

ASSIGNOR : Redox Technologies
Inc.

TITLE : PREPARATION OF DIALKYL PEROXIDES

DECLARATION OF JOHN KOLLAR

JOHN KOLLAR, the undersigned, deposes and says that:

1. I am a chemist having obtained an A.B. degree in chemistry from Columbia College in 1957. I have also completed graduate course work in chemistry in 1960 at The Carnegie Institute of Technology. I am currently the president and majority shareholder in Redox Technologies Inc. a research and consulting firm specializing in chemicals, fuel and energy. I have specialized expertise in oxidation chemistry, especially that which is directed toward large volume commercial chemicals. During the course of my career, I have obtained approximately 60 patents issued in the United States and approximately 1000 patents issued worldwide.

2. I believe myself to be the original, first and sole inventor of a process for the preparation of a dialkyl peroxide comprising reacting one or more members selected from the group consisting of an alkylating alcohol of the formula ROH, and an olefin of the formula $(R^2)(R^{2a})C=C(R^3)(R^{3a})$, wherein R is C_1 - C_{10} alkyl, and R^2 , R^{2a} , R^3 , and R^{3a} are independently selected from hydrogen and C_1 - C_{10} alkyl;

with a hydroperoxide of the formula R^1OOH , wherein R^1 is C_1 - C_{10} alkyl; in the presence of an effective amount of a substantially solid, insoluble, heterogenous acidic catalyst; followed by separation of the reaction mixture from said catalyst; and more particularly solid acid catalyzed, especially at least 10% cross-linked ion exchange resin catalyzed, commercially viable production of DtBP (di-*tertiary* butyl peroxide) from *t*BHP (*t*-butyl hydroperoxide) and isobutylene and/or *t*BA (*tertiary* butyl alcohol) [hereafter "the DtBP invention"].

3. The need for commercial production of DtBP resulted from my earlier work which established its commercial use as both a stoichiometric reactant with methanol to produce EG (ethylene glycol) by dehydrodimerization, and by less than stoichiometric DtBP free radical induced addition of methanol to formaldehyde to produce EG. These methods of producing EG which utilize DtBP are patented in the United States as Patent Nos, 4,337,371; 4,393,252; 4,412,084; and 4,412,085.

4. The DtBP invention was reduced to practice by myself with the assistance of my son J. Christopher Kollar who, acting under my direction, conducted experiments designed by me in the field of methanol based EG production. The first reaction of *iso*-butylene with *t*BHP in the presence of an effective amount of a substantially solid, insoluble, heterogenous acidic catalyst, occurred in the presence of a toluene diluent at 60° C on June 29, 1979, as evidenced by the Declaration of John Christopher Kollar, attached as Exhibit "1". I carried out an analysis of the reaction products which confirmed that DtBP had been produced, which I then noted on the laboratory notebook record of the reaction kept by J. Christopher Kollar, attached to his Declaration as Exhibit "1-a", and which I also communicated to him verbally.

5. A further embodiment of the DtBP invention comprising reacting isobutylene with *t*BHP without diluents at a high degree of conversion was conducted by myself on July 27, 1979 as evidenced by the attached Exhibit "2".

6. I have, to the extent of the resources available to me, pursued the development of this major technology diligently and vigorously on a priority basis.

7. I had conceived and reduced to practice, at least as early as 1979, the pure isobutylene reaction with pure *t*BHP as one of many component parts necessary to establish a complete and technologically accessible methanol-based EG manufacturing process. At this time the commercial aspects of the DtBP invention were recognized by me and incorporated in a Redox EG Process disclosure agreement between Redox and Celanese Corp. This disclosure agreement, which was negotiated with Celanese personnel at their R&D center in Corpus Christi, Texas, carried with it an obligation of confidentiality, and was signed on November 2, 1979. Portions of that document, attached hereto as Exhibit "3", constitute a written and corroborated record of embodiments within the scope of my conception of the DtBP invention not actually reduced to practice at that time.

8. The first inclusion of a description of my DtBP invention in a formal Celanese document was in a Report entitled "Redox II Design and Economics Update", which was co-authored by Dennis D. Dickinson and William N. Uber, and is dated September 4, 1981. A part of this Report, consisting of a distribution page, a title page, a signature page and six pages describing various aspects of commercial *t*BHP alkylation to DtBP are attached hereto as Exhibit "4".

9. I reduced to practice further embodiments of my DtBP invention in late 1982. As part of an ongoing cooperative effort between Redox and Celanese at that time, an alternative optimization of the basic process, using alcohol by-products produced downstream (*t*-butanol and/or mixtures of *t*-butanol and methanol and ethanol) was used to alkylate *t*BHP to DtBP. This was disclosed to Celanese personnel on January 21, 1983. The summary report presented to Celanese is attached hereto as Exhibit "5".

10. As part of an effort to obtain development funding, I disclosed to personnel of Arco Chemical Company (ACC), under the provisions of a confidentiality agreement, the Redox EG Process (as described in Paragraphs 7. -9. above) on May 14, 1987 in Newton Square, PA. That disclosure included documents which contained a description of the DtBP invention. The pertinent parts of the Redox EG disclosure to ACC are attached hereto as Exhibit "6", and will be observed to incorporate therein at least a part of each of Exhibits "3" through "5" inclusive.

11. All statements made herein of my own knowledge are true and all statements made on information and belief are believed to be true; and further, these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the above-identified application or any patent issued thereon.

4-20-96

DATE

John Kollar
JOHN KOLLAR